



Ratio of $^{36}\text{Cl}/\text{Cl}$ in ground ice of east Siberia and its application for chronometry

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[1] Abundance of the cosmogenic nuclide chlorine-36 (^{36}Cl) was measured together with the chloride (Cl^-) concentration in different horizons of Quaternary permafrost samples collected from various types of ground ice in the northeastern part of Siberia. The $^{36}\text{Cl}/\text{Cl}$ in 32 samples ranged in value from 2.4×10^{-14} to 1.4×10^{-12} . Nonetheless, after a few extreme values were excluded, these $^{36}\text{Cl}/\text{Cl}$ ratios provided a local permafrost chronometry. The general concordance of the modeled ages with geological expectations and other chronological methods supports the potential power of the proposed dating method. However,

the large observed change in ratios from higher to lower values during the transition from Last Glacial Maximum to Holocene climatic conditions remains unexplained. An attempt to make use of the corresponding beryllium-10 (¹⁰Be) absolute concentrations in the same samples failed because input of ¹⁰Be attached to particulate matter into permafrost is unknown. Further ³⁶Cl/Cl serial measurements of modern precipitation and fossil ground ice are needed to refine this dating method into a practical tool with a clear protocol.

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1. Introduction

[2] Permafrost is a paleoenvironmental archive that contains several proxy records such as fossil faunal, floral, and microbial communities and greenhouse gases that existed before human environmental impact. In the Eurasian Arctic, ancient frozen ground exists because of the persistent harsh, cold climatic conditions. Permafrost underlies most of the Siberian territory; however, the question of whether it has existed permanently since its first occurrence remains unresolved. The oldest signs of permafrost conditions are related to frost crack pseudomorphs in late Pliocene deposits at the Krestovka River in the Kolyma lowlands [Sher, 1971, 1974] and in the Val'karai lowlands on the northern coast of Chukotka [Arkhangelov *et al.*, 1985]. The paleoecological data of Sher [1997] indicated that most ancient permafrost records are about 3 million years old.

[3] The potential of permafrost as a paleoenvironmental archive of geochemical, cryolithological, and biological characteristics with ages that correspond to those of permafrost makes the development of an accurate permafrost chronology an essential objective of geological, climatological, and paleoecological studies. Permafrost chronology is important for outlining the stratigraphy and Quaternary history of the permafrost region as well

as for detecting the temporal vertical dynamics of the permafrost.

[4] Previous paleoenvironmental studies in Arctic permafrost have used different chronological methods such as radiocarbon dating [e.g., Mackay *et al.*, 1972; Moorman *et al.*, 1996; Murton *et al.*, 1997; Sulerzhitsky and Romanenko, 1997; Vasil'chuk and Vasil'chuk, 1998; Vasil'chuk *et al.*, 2000; Schirmer *et al.*, 2002a; Grosse *et al.*, 2007], luminescence dating [e.g., Krbetschek *et al.*, 2000; Andreev *et al.*, 2004, 2009], and the ²³⁰Th/U disequilibria method [e.g., Schirmer *et al.*, 2002b, 2003; Wetterich *et al.*, 2008a]. Using these dating methods, only the age of sedimentation and of organic remains within permafrost were determined; the age of the permafrost aggradation, especially for early Pleistocene to late Pleistocene deposits, could not be determined. Continuous chronologies of permafrost do not extend beyond the limit of radiocarbon, and there are but few reports with indirect dates of mid-Pleistocene permafrost [e.g., Froese *et al.*, 2008]. The recently proposed geochronology method using the long-lived cosmogenic radionuclide ³⁶Cl [Gilichinsky *et al.*, 2007] could potentially extend the dating range beyond several hundred thousand years; in so doing, this method would find a wide variety of applications in Quaternary paleoenvironmental and paleoecological reconstructions, e.g., estimating the time scale for the long-term preservation of fossils, tracing the history of greenhouse gases, and

determining the age of inclusions of viable microorganisms adapted to a permanently frozen environment.

[5] Common ground ice features within the permafrost are ice wedges formed by annually repeated contraction cracking of upper frozen ground layers. The precipitation that falls in winter enters the ice wedge frost crack as meltwater during melt season and refreezes there, forming a new vertical ice vein and subsequently growing ice wedges. Two main types of ice wedges are distinguished: syngenetic ice wedges form synchronously with accumulation and freezing of the surrounding sediments; epigenetic ice wedges form during freezing of already existing sediments [van Everdingen, 1998]. Depending mainly on the temperature regime, the water supply, and the substrate, other ground ice types such as segregation ice, pore ice, or buried ice bodies of various structure and extent are also substantial parts of permafrost.

[6] The global chlorine cycle has been intensively studied, mostly in connection with atmospheric ozone depletion [Graedel and Keene, 1996]. Most stable chlorine enters the troposphere in sea spray. Volcanoes add not more than 15–20% to the tropospheric chlorine injection in the form of HCl [Graedel and Keene, 1996]; this source is highly sporadic and unevenly distributed on the surface. The addition of dust-borne chlorine from the surface is usually neglected in the contemporary atmospheric budget. Under standard atmospheric conditions, the chlorine atoms produced occur as Cl⁻ due to this element's strongly hydrophilic nature and high electron affinity. This is advantageous for many environmental and hydrological applications because chloride acts as a conservative tracer and undergoes minimal chemical interactions in the environment. In the course of its geochemical cycle, chlorine is removed from the atmosphere by wet and, to a smaller extent, by dry precipitation. The deposition flux depends on the distance from the oceans and on wind direction and speed [Johnston and McDermott, 2008].

[7] Under contemporary conditions, the Siberian regions west of 140°E receive most of the moisture from the relatively warm northern Atlantic Ocean as indicated by the west to east direction of the main moisture fluxes in northern Eurasia [Kuznetsova, 1998]. In contrast, moisture transport from the cold, predominantly ice-covered Arctic Ocean to Siberia is considered to be negligible. Therefore, the varying distance of northern Siberian regions from the Arctic Ocean is not expected to influence

the deposition flux of stable chlorine. During ice wedge formation, the stable chlorine contained in the precipitation is incorporated into the ice.

[8] The cosmogenic radionuclide ³⁶Cl has a half-life of 301,000 years, and is produced in the Earth's atmosphere by nuclear reactions of cosmic rays (CR) with atoms of air. The primary CR penetrating the atmosphere generate secondary cascades which develop in the downward direction, so the stratosphere yields about 55% of the mean atmospheric production of ³⁶Cl and the troposphere contributes the remaining 45% [Masarik and Beer, 1999]. The dominant contribution comes from argon-40 (⁴⁰Ar) spallation, with a minor addition from neutron activation of ³⁶Ar and ³⁵Cl. According to calculations, the mean global atmospheric production rate of cosmogenic ³⁶Cl is close to 20 atoms m⁻²s⁻¹. The production rate has a strong latitudinal dependence caused by the shielding effect of the Earth's dipole magnetic field. However, most of this effect is eliminated by strong mixing of the stratospheric air masses [Heikkila et al., 2008].

[9] The production of ³⁶Cl, as well as other cosmogenic nuclides, is affected by temporal changes in solar magnetic activity, which modulates the CR flux via the solar wind. A stronger solar wind deflects more CR away from Earth decreasing ³⁶Cl production, and vice versa. The 11-year cycle and hundred-year-long intervals of depressed solar activity are apparent in annually resolved radionuclide records [Muscheler et al., 2007]; however, these cycles are negligible when averaged on the geological time scale. Long-term variations of the ³⁶Cl production rate on multimillennial time scales are caused by slow changes of the geomagnetic field intensity [Baumgartner et al., 1998].

[10] ³⁶Cl that has been produced and circulated in the stratosphere is preferentially transported to the troposphere at midlatitudes, in the regions of the polar and subpolar jet streams, causing maximum ³⁶Cl fallout between 30° and 60° in both hemispheres [Lal and Peters, 1967; Heikkila et al., 2008]. After a residence time of about one week, ³⁶Cl is then removed from the troposphere by wet and dry deposition [Bentley et al., 1986]. Its local surface flux is strongly influenced by the precipitation rate, and corrections should be applied to account for this effect [Blinov et al., 2000]. Obviously, the atmospheric air transport is influenced by climatic, as well as meteorological conditions [Heikkila et al., 2008]. Thus, temporal and spatial variations seen in the ³⁶Cl local surface flux reflect

a combination of variations in production, transport, and depositional processes. Recently, new surface precipitation measurements have estimated the mean fallout rate of ³⁶Cl in European midlatitudes (30–70°N) as 43.7 atoms m⁻²s⁻¹ [Johnston and McDermott, 2008].

[11] The mean lifetime of ³⁶Cl is $\tau_{36} = T_{1/2}/\ln 2 = 434,000$ years. After the meteoric component following the surface water flow is incorporated into the permafrost, the ratio of ³⁶Cl to stable chlorine decreases with time by the process of exponential radioactive decay:

$$\frac{d\left(\frac{{}^{36}\text{Cl}}{\text{Cl}}\right)}{\frac{{}^{36}\text{Cl}}{\text{Cl}}} = -\frac{dt}{\tau_{36}}. \quad (1)$$

It was assumed [Gilichinsky *et al.*, 2007] that after an ice wedge system closed, the time interval Δt since formation of its horizons could be calculated according to (1) from the corresponding ³⁶Cl/Cl ratios:

$$\Delta t = t_2 - t_1 = \tau_{36} \cdot \ln\left(\frac{{}^{36}\text{Cl}/\text{Cl}(t_1)}{{}^{36}\text{Cl}/\text{Cl}(t_2)}\right). \quad (2)$$

For a surface base sample of zero age, equation (2) gives the absolute age of the deeper sample. The precondition of the proposed method's validity is the constant surface ³⁶Cl/Cl over Δt .

[12] Groundwater ages in the range of up to one million years established on the basis of ³⁶Cl concentration measurements have been reported [Bentley *et al.*, 1986]. It is expected that syngenetic permafrost ice samples with ages up to 1–2 million years can be dated with a similar procedure. The ³⁶Cl/Cl method was tested on the late and mid-Pleistocene syngenetic permafrost ice wedges from Cape Svyatoy Nos and Bol'shoy Lyakhovsky Island, resulting in an age range of 460 ± 130 thousand years for regional Pleistocene permafrost, a range that was supported by indirect paleontological and geochronological data [Gilichinsky *et al.*, 2007]. This work, which applied the ³⁶Cl/Cl method to additional permafrost samples from various stratigraphical units from the same region, mainly aimed to confirm the principles and improve the database.

2. Sampling of Ground Ice

[13] Quaternary syngenetic permafrost deposits with two levels of large ice wedges exist in the western part of the East Siberian Sea along the

Dmitry Laptev Strait; these deposits are located on both the west and north coasts of Cape Svyatoy Nos and Oyogos Yar, and on the southern coast of Bol'shoy Lyakhovsky Island. A special sampling campaign was organized in this region during the summer seasons from 2004 to 2007 (Figure 1). The samples of ice wedges (Figure 2) were extracted either from the boreholes, or from outcrops with well-defined horizon structure. A summary of the ice wedge data appears in Table 1. The majority of the samples come from several generations of ice wedges. A few samples consist of segregation ice or buried ice lenses.

[14] Permafrost samples (Table 1, samples 1–4) from the cross section of Cape Svyatoy Nos (site A in Figures 1 and 2) were delivered for dating in summer of 2004. This section included two different systems of syngenetic ice wedges formed during the mid-Pleistocene (Yukagir Suite) and late Pleistocene (Yedomo Suite). The sample (Table 1, sample 5) of a middle Pleistocene syngenetic ice wedge (Yukagir Suite) from Bol'shoy Lyakhovsky Island was collected in order to confirm the dating result from Cape Svyatoy Nos.

[15] Sixteen ice wedge samples (Table 1, samples 6–21) were collected from cliff exposures on both coasts of Dmitry Laptev Strait in 2007. Ten samples (Table 1, samples 6–15) were collected at the south coast of Bol'shoy Lyakhovsky Island (site B in Figures 1 and 2). Six samples were taken at the coast of Oyogos Yar (site C in Figures 1 and 2). The studied sequences cover the Saalian Ice Complex deposits of the Yukagir Suite (Table 1, samples 5, 6, and 8), pre-Eemian epigenetic ice wedges in fine-grained floodplain deposits of the Kuchchugui Suite (Table 1, samples 7 and 12), probable Early Weichselian syngenetic ice wedges of the Bychchaguy Suite (Table 1, samples 14, 18, and 19), huge syngenetic ice wedges of the late Pleistocene Yedomo Suite (Table 1, samples 10, 13, 15, 16, and 21), and young syngenetic ice wedges formed in Holocene thermokarst depressions (Table 1, samples 9, 11, 17, and 20). The exposed sequences were surveyed, described, photographed, and sketched according to sediment structures and cryostratigraphies [Schirrmeyer *et al.*, 2008; Opel *et al.*, 2009; Wetterich *et al.*, 2009]. Most ice wedge samples were taken with an axe; one of them (Table 1, sample 8) was obtained with a chain saw.

[16] In 2006, five samples of ground ice (Table 1, samples 22–26) were collected at Khaptashinsky Yar on the right bank of the Khroma Bay of the East Siberian Sea (site D in Figures 1 and 2). The

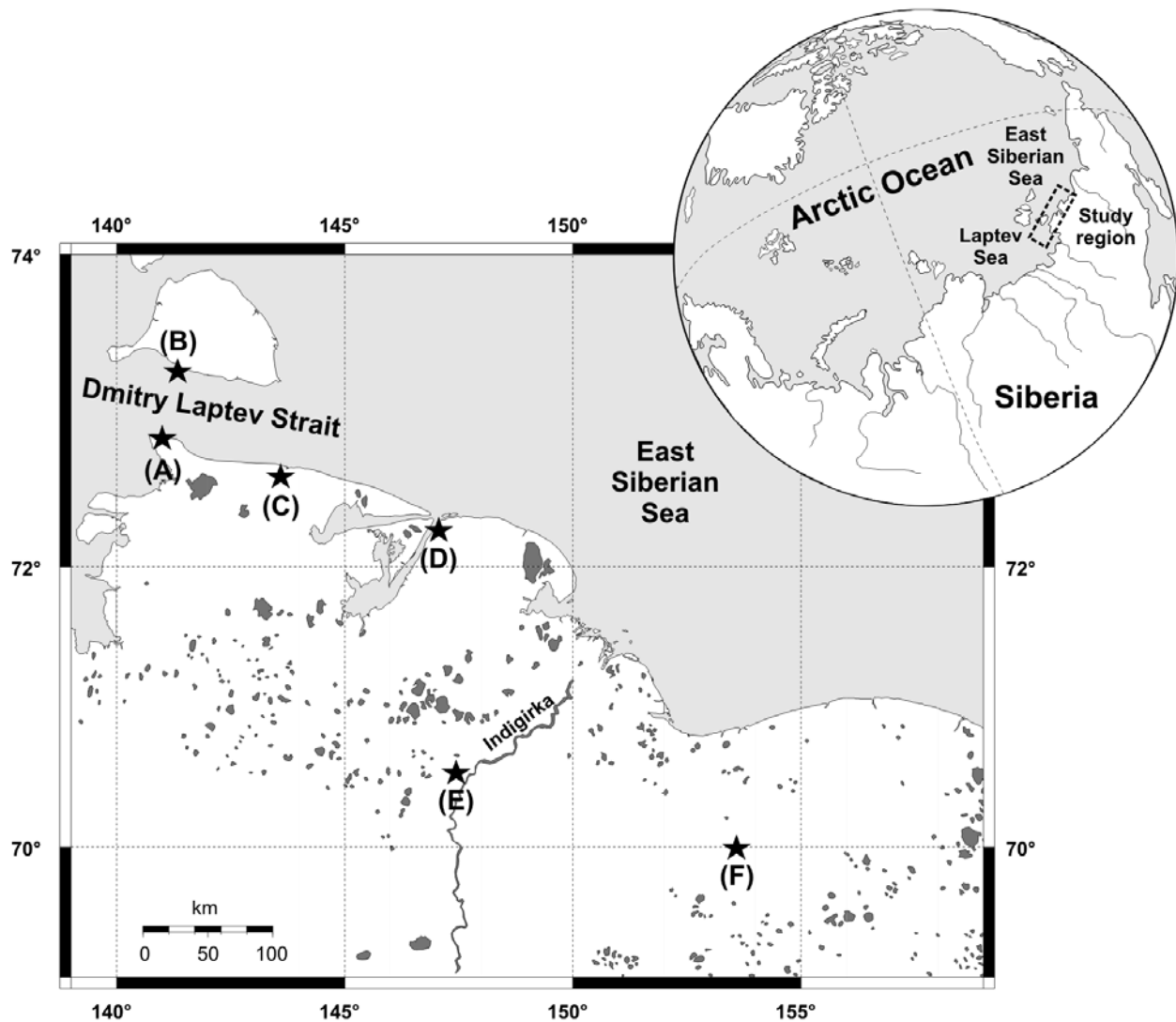


Figure 1. Location of sampling sites in northeast Siberia: site A, Cape Svyatoy Nos; site B, Bol'shoy Lyakhovsky Island; site C, Oyogos Yar coast; site D, Cape Khaptashinsky Yar; site E, Allaikha River; site F, Bol'shoy Khomus Yuryakh River.

first three samples (Table 1, samples 22–24) were taken near the sea shore from different depths in the same borehole. They belong to a syngenetic ice wedge of the Yedoma Suite. Sample 25 was collected from another borehole at the same location. It belongs to a syngenetic ice wedge of the middle Pleistocene Ice Complex. Sample 26 was taken from an ice lens of a nearby river section. This exposure presumably corresponds to the same middle Pleistocene Ice Complex as sample 25, and the quality of the sample was marked as poor.

[17] Two samples (Table 1, samples 27 and 28) were collected on the banks of Allaikha River, the left tributary of the Indigirka River at a distance of about 100 km from the Arctic Ocean (site E in Figures 1 and 2). Sample 27 was taken from a

borehole and belongs to a syngenetic ice wedge of the late Pleistocene Ice Complex (Yedoma Suite). Sample 28 was cut from the middle Pleistocene Ice Complex (Achagy Suite) exposure on the river bank section with an estimated age of more than 300 thousand years. This was the only sample of segregated ice cement finally obtained by melting out the frozen permafrost sample.

[18] Four samples (Table 1, samples 29–32) were obtained in the Kolyma lowlands in 2005, in the upper reaches of the Bol'shoy Khomus Yuryakh River (site F in Figures 1 and 2). They represent two middle Pleistocene horizons. The upper horizon is represented by three samples. Two samples of segregated ice (Table 1, samples 29 and 30) melted from a frozen deposit that may be 300–

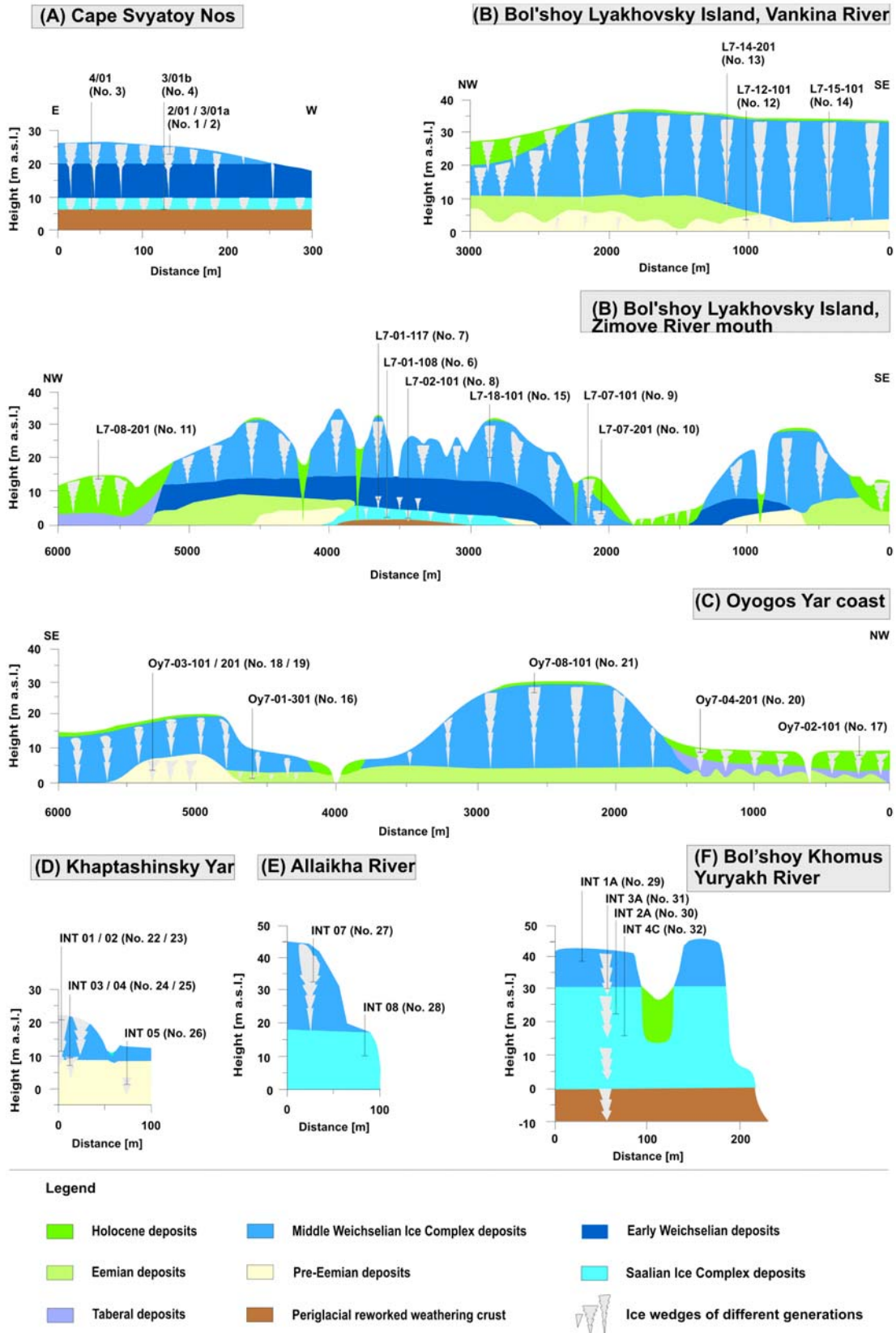


Figure 2. Generalized overview schemes of the studied exposures: site A, Cape Svyatoy Nos; site B, Bol'shoy Lyakhovsky Island; site C, Oyogos Yar coast; site D, Cape Khaptashinsky Yar; site E, Allaikha River; site F, Bol'shoy Khomus Yuryakh River.

Table 1. Field Data, Analytical Results, and Calculated Ages^a

Sample Number	Sample ID	Sample Type	Sample Depth Below Surface (m b.s.)	Expected Age (10^3 Years)	Local Stratigraphy	N (Cl^-) (ppm)	N ($^{36}\text{Cl}/\text{Cl}^-$) (10^{-12})	N(^{36}Cl) (10^4 at. g^{-1})	$^{36}\text{Cl}/\text{Cl}$ Age (10^3 Years)
<i>Cape Svyatoy Nos (2004), 72°51'N, 141°05'E</i>									
1	2/01	IW	3.35–4.35	17–25	Yedoma ^b	7.2 ± 0.6	0.25 ± 0.06	3.06 ± 0.78	34 (base)
2	3/01a	IW	4.35–5.1	25–50	Yedoma ^b	6.2 ± 0.5	0.37 ± 0.08	3.9 ± 0.9	34 (base)
3	4/01	IW	7.85–8.4	200–400	Yukagir ^b	50.8 ± 1.2	0.11 ± 0.02	9.49 ± 2.08	484 ± 161
4	3/01b	IW	20.5–20.7	200–400	Yukagir ^b	27.1 ± 0.9	0.088 ± 0.018	4.05 ± 0.84	581 ± 166
<i>Bol'shoy Lyakhovsky Island (2005), 73°17'N, 141°20'E</i>									
5	PFBLO5	IW	25	200–400	Yukagir	8.1 ± 0.5	0.13 ± 0.008	1.79 ± 0.16	341 ± 43
<i>Bol'shoy Lyakhovsky Island (2007), 73°17'N, 141°20'E</i>									
6	L7-01-108	IW	28	>150	Yukagir ^c	3.8 ± 0.3	0.14 ± 0.007	0.90 ± 0.08	309 ± 40
7	L7-01-117	IW	27	130–150	Kuchchugui ^c	7.4 ± 0.3	0.08 ± 0.003	1.01 ± 0.06	552 ± 37
8	L7-02-101	IW	27	>150	Yukagir ^c	7.3 ± 0.3	0.13 ± 0.008	1.61 ± 0.12	341 ± 43
9	L7-07-101	IW	3	1–10	Holocene ^d	10.4 ± 0.4	0.23 ± 0.008	4.06 ± 0.21	<11
10	L7-07-201	IW	4	30–50	Yedoma ^d	6.2 ± 0.3	0.23 ± 0.008	2.42 ± 0.14	40 (base)
11	L7-08-201	IW	1.9	1–10	Holocene ^{d,e}	14.2 ± 0.5	0.19 ± 0.01	4.58 ± 0.29	<11
12	L7-12-101	IW	28	130–150	Kuchchugui ^c	10.3 ± 0.4	0.07 ± 0.004	1.22 ± 0.08	609 ± 42
13	L7-14-201	IW	22	40–50	Yedoma ^{d,e}	4.1 ± 0.2	0.09 ± 0.005	0.63 ± 0.05	500 ± 41
14	L7-15-101	IW	11	100–120	Bychchagy ^c	3.5 ± 0.2	0.89 ± 0.005	5.29 ± 0.30	failed
15	L7-18-101	IW	13.2	30–50	Yedoma ^d	3.1 ± 0.2	0.29 ± 0.018	1.53 ± 0.14	40 (base)
<i>Oyogos Yar coast (2007), 72°36'N, 143°36'E</i>									
16	Oy7-01-301	IW	1	30–50	Yedoma	6.1 ± 0.3	0.109 ± 0.007	1.13 ± 0.13	40 (base)
17	Oy7-02-101	IW	2	1–10	Holocene ^f	20.1 ± 0.4	0.031 ± 0.003	1.06 ± 0.15	<11
18	Oy7-03-101	IW	15	100–120	Bychchagy	4.0 ± 0.2	0.30 ± 0.01	2.04 ± 0.21	68 ± 31
19	Oy7-03-201	IW	15	100–120	Bychchagy	4.0 ± 0.2	0.28 ± 0.01	1.90 ± 0.20	98 ± 31
20	Oy7-04-201	IW	1.5	1–10	Holocene ^f	9.4 ± 0.2	0.032 ± 0.003	0.51 ± 0.07	<11
21	Oy7-08-101	IW	3.5	30–50	Yedoma	4.4 ± 0.2	0.52 ± 0.02	3.89 ± 0.38	40 (base)
<i>Khaptashinsky Yar (2006), 72°14'N, 147°03'E</i>									
22	INT 01	IW	0.5–2	20–30	Yedoma	5.9 ± 0.3	0.087 ± 0.012	0.87 ± 0.13	25 (base)
23	INT 02	IW	8–9	20–30	Yedoma	3.4 ± 0.2	0.38 ± 0.02	2.19 ± 0.17	25 (base)
24	INT 03	IW	11.5–12.2	20–30	Yedoma	7.1 ± 0.3	0.29 ± 0.02	3.50 ± 0.28	25 (base)
25	INT 04	IW	12.3–15.3	200–300	Khroma ^g	9.8 ± 0.4	0.22 ± 0.01	3.66 ± 0.22	80 ± 56
26	INT 05	SI	>15.3	>300	Khroma ^g	83.8 ± 0.8	0.11 ± 0.01	15.65 ± 1.71	381 ± 65
<i>Allaikha River (2006), 70°34'N, 147°27'E</i>									
27	INT 07	IW	6–8	20–30	Yedoma ^g	7.4 ± 0.3	0.29 ± 0.02	3.64 ± 0.29	25 (base)
28	INT 08	SI	10.3–11.2	300–500	Achagy ^g	38.2 ± 0.6	0.19 ± 0.014	12.32 ± 0.93	209 ± 44
<i>Bol'shoy Khomus Yuryakh River (2005), 70°00'N, 153°37'E</i>									
29	INT 1A	SI	1.4–5.4	200–400	–	6.2 ± 0.3	0.34 ± 0.04	3.58 ± 0.46	300 (base)
30	INT 2A	SI	0.7–2.7	200–400	–	6.8 ± 0.3	0.31 ± 0.02	3.58 ± 0.28	300 (base)
31	INT 3A	IW	12	200–400	–	7.7 ± 0.4	0.024 ± 0.005	0.31 ± 0.07	<11
32	INT 4C	SI	8.4–12	500–600	–	12.7 ± 0.4	0.21 ± 0.02	4.53 ± 0.45	496 ± 78
<i>Lower Kolyma River (2004, 2005), 68°45'N, 161°20'E</i>									
33	INT-2004-S	snow	surface	zero age	modern	0.7 ± 0.1	0.012 ± 0.002	0.014 ± 0.003	<11
34	INT-2005-S	snow	surface	zero age	modern	1.3 ± 0.2	0.014 ± 0.001	0.030 ± 0.005	<11
<i>Yana River (2006), 67°41'N, 135°44'E</i>									
35	INT-10W	river water	surface	zero age	modern	0.5 ± 0.1	0.39 ± 0.02	0.33 ± 0.067	<11

^aThe sample type is indicated as ice wedge (IW) or segregation ice (SI).

^bGilichinsky et al. [2007].

^cAndreev et al. [2004].

^dAndreev et al. [2009].

^eWetterich et al. [2009].

^fOpel et al. [2009].

^gKaplina [1981].

400 thousand years old, according to paleontological evidence. The third sample (Table 1, sample 31) was cut out of a syngenetic ice wedge from the same horizon. The fourth sample (Table 1, sample 32) was collected from a nearby borehole and also consists of melted, segregated ice from permafrost

deposits of a lower horizon with an age of about 500–600 thousand years.

[19] In addition, three samples from modern precipitation and surface water were collected from a nearby geographical region to serve as references

(Table 1, samples 33–35). Two snow samples were collected in the vicinity of the Kolyma River's lower reach at the beginning of springtime in 2004 and 2005. They represent the contemporary concentration of chemical elements and cosmogenic nuclides in winter precipitation, although they could not be attributed to a specific time interval and could be subject to local conditions such as wind advection and mixing. One water sample was collected from the Yana River in summer 2006.

3. Methods

[20] During the field stage a volume of ~ 2 L of the initial sample was melted, poured into a clean plastic bottle, marked, and shipped to a laboratory for follow-up studies. The experimental procedure for samples 1–4 was described by *Gilichinsky et al.* [2007]. Chloride concentrations were measured on filtered aliquots using ion chromatography (IC, Dionex DX-320); samples 6–21 were measured at the Alfred Wegener Institute for Polar and Marine Research Potsdam (AWI, Germany), and samples 5 and 22–35 were measured at the Swiss Federal Institute of Aquatic Science and Technology (Eawag, Switzerland). Afterward, the samples for ^{36}Cl measurements were prepared at Eawag following the standard method [*Synal et al.*, 1994]. One sample only (Table 1, sample 11) required the addition of 3.8 mg Cl carrier solution to increase the total chlorine content. Measurements of $^{36}\text{Cl}/\text{Cl}$ ratios were carried out by Accelerator Mass Spectrometry (AMS) at the Swiss Federal Institute of Technology facility (ETH Zurich, Switzerland).

4. Results

4.1. Concentrations and Ratio

[21] Measured chloride concentrations and $^{36}\text{Cl}/\text{Cl}$ for the ice samples are summarized in Table 1. The results for the first four samples were already presented by *Gilichinsky et al.* [2007]. The chloride concentrations range from 3 ppm, typical of continental rainwater, up to 84 ppm which exceeds even the mean groundwater concentration. Note that Holocene ice wedges from Bol'shoy Lyakhovskiy Island and Oyogos Yar exhibit significantly higher Cl^- concentration than those of the Late Pleistocene (Yedoma Suite), possibly related to sea spray effects due to the Holocene transgression; uncertainties in the Cl^- concentration measurements were systematically less than 10%. The $^{36}\text{Cl}/\text{Cl}$ ratios range from 2.4×10^{-14} to $3.9 \times$

10^{-13} for ice; uncertainties in $^{36}\text{Cl}/\text{Cl}$ ratios were dominated by an average relative error of about 12% in counting the rare ^{36}Cl isotope. The chloride concentration and $^{36}\text{Cl}/\text{Cl}$ ratios in permafrost both exceed the values measured for modern snow and river water (Table 1, samples 33–35). For comparison, values of $0.02\text{--}0.5 \times 10^{-12}$ were measured for $^{36}\text{Cl}/\text{Cl}$ in modern rainwater collected in Europe [*Johnston and McDermott*, 2008].

[22] ^{36}Cl concentrations do not show any clear dependence on chloride concentrations, but rather appear to depend on changing atmospheric transport and climatic conditions. The scatter in the data presumably reflects complicated dynamics of ^{36}Cl fallout and local fixation conditions in ice. The main feature of the calculated concentrations is their general excess, by a factor of about 20, over the values known for the Holocene-related part of Greenland ice cores that directly reflect atmospheric production by CR. Apparently these ^{36}Cl concentrations do not reflect direct freezing out of surface water containing a constant meteoritic value. At the same time, any contamination of the deeper sediment horizons by recent bomb-produced surface radioactivity could be excluded by the depth profile of the results.

[23] The expected geologic age of the permafrost samples (Table 1, column 5) corresponds to a wide time interval. The $^{36}\text{Cl}/\text{Cl}$ scattering demands an additional explanation. Arctic ice cores could have provided further data on a comparable time scale, but because of the very low chlorine content, the $^{36}\text{Cl}/\text{Cl}$ was not systematically measured there. The ^{36}Cl concentration in the GRIP ice core [*Wagner*, 1999] decreased by a factor of 2 to 2.5 at the transition from the Last Glacial Maximum to the Holocene. However, this decrease was reasonably explained by the corresponding increase in the precipitation rate which diluted the cosmogenic ^{36}Cl in the ice volume. In our analysis we have assumed that the $^{36}\text{Cl}/\text{Cl}$ ratio, naturally averaged over a thousand-year interval of constant climatic conditions and fixed in the permafrost ice, was insensitive to changes in the supply of surface water. However, additional sources of chlorine, like eroded dust or sea-salt aerosol input, may have altered the local balance. Similar to the GRIP record, the transition from late Pleistocene (Yedoma Suite) to Holocene is reflected in Siberian ground ice as a change of $^{36}\text{Cl}/\text{Cl}$ from $(2.5\text{--}3.5) \times 10^{-13}$ to $(1.5\text{--}2.5) \times 10^{-14}$, except for samples from Bol'shoy Lyakhovskiy Island (Table 1, column 8, samples 9 and 11). This discrepancy was

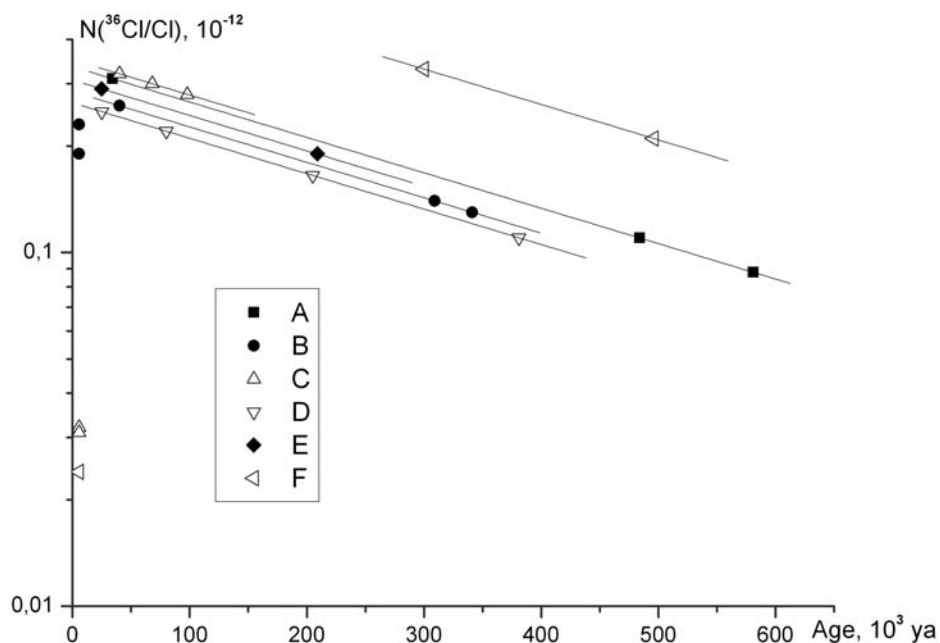


Figure 3. Local permafrost chronologies: site A, Cape Svyatoy Nos; site B, Bol'shoi Lyakhovskiy Island; site C, Oyogos Yar coast; site D, Khaptashinsky Yar; site E, Allaikha River; site F, Bol'shoi Khomus Yuryakh River.

probably caused by local peculiarities of thermokarst-related processes at Bol'shoi Lyakhovskiy Island.

[24] The excess of ³⁶Cl in older samples could be a result of in situ production. Some in situ ³⁶Cl produced in ice wedges after deposition is also stored in permafrost together with meteoric ³⁶Cl. However, according to our calculations [Tikhomirov and Blinov, 2009] the buildup of in situ ³⁶Cl is negligible in late Pleistocene and Holocene Siberian permafrost, and is unlikely to be important for any permafrost horizons younger than one million years.

[25] The ¹⁰Be concentrations were measured in sixteen of the studied samples. The long-lived cosmogenic radionuclide ¹⁰Be was initially considered a supplementary source of information about the transport of surface water to the ice wedges. In modern water samples (snow, river water) its concentration was measured at about 10⁴ at. g⁻¹, close to the value for Greenland ice cores or for rainwater collections. In contrast, the concentration of ¹⁰Be in permafrost samples showed values extending from 10⁵ up to 10⁷ at. g⁻¹ with a ³⁶Cl/¹⁰Be ratio as low as 0.001, excluding an atmospheric origin. The excess may represent an influx of ¹⁰Be to permafrost ice with dust from eroded rock material. The fine particulate component was present in all studied ice samples and the

division of ¹⁰Be between particles and water is unknown. After this fact had become clear, measurements of ¹⁰Be concentrations in the next samples were canceled.

4.2. Age Determination

[26] The ³⁶Cl/Cl ratios of the ice wedges were used for permafrost age determination. Ground ice samples with a ³⁶Cl/Cl ratio below 5 × 10⁻¹⁴ belonging to the Holocene period defined as <11 ka were excluded from the following procedure because their initial ³⁶Cl/Cl clearly differs from that of the older samples. For all study sites, we assumed the age of the samples representing the youngest late Pleistocene horizon (Yedoma Suite) according to stratigraphy classification and geological interpretation (Table 1, column 5). The resulting ages were used as the base age for each local chronology. The ages of the middle Pleistocene samples were calculated as the sum of the median base age and the time interval was determined according to (2). The results are given in Table 1, column 10, and are illustrated by lines of local permafrost chronologies in Figure 3. The age determination errors were calculated as a statistical sum which included measurement errors of the ³⁶Cl/Cl ratio and the stable Cl concentration. Geological uncertainties for base samples were not included in the errors because of their nonstatistical nature. As noted above, the ³⁶Cl/Cl ratios of Holocene samples do

not lay on the decay curves because of the change from higher to lower ratio values during the transition from late Pleistocene to Holocene. Elucidating the reasons for the changes in the $^{36}\text{Cl}/\text{Cl}$ ratio during the Holocene will require additional research.

4.3. Discussion

[27] Stratigraphic ages coincide with those calculated from $^{36}\text{Cl}/\text{Cl}$ for middle Pleistocene (Late Saalian) ground ice at the Khomus Yuryakh River, Khaptashinsky Yar, Bol'shoy Lyakhovsky Island, and Cape Svyatoy Nos study sites as well as for late Pleistocene (Early Weichselian) ground ice on the Oyogos Yar coast. These results confirm the general utility of ground ice dating with $^{36}\text{Cl}/\text{Cl}$ for northeast Siberian permafrost. The age for middle Pleistocene ground ice at the Allaikha River site was younger than expected, although close enough considering the related uncertainties.

[28] Four samples from Bol'shoy Lyakhovsky Island (Table 1, samples 7 and 12–14) are omitted from the general interpretation since their stratigraphy (site B in Figure 2) strongly contradicts the calculated ages. According to the stratigraphy, samples 13 and 14 are connected to adjacent younger post-Eemian horizons and could not therefore have been formed some hundred thousand years ago. Corresponding peat deposits from sample 13 were independently dated by the radiocarbon method to $42.61 \pm 1.12/-0.98$ ka BP. The ice wedge sample 14 showed the highest $^{36}\text{Cl}/\text{Cl}$ ratio, more than twice the mean of the other middle Pleistocene samples; it was also characterized by one of the highest absolute concentrations of ^{36}Cl . The $^{36}\text{Cl}/\text{Cl}$ ratio was practically identical in the remaining exceptional samples (samples 7, 12, and 13). Two of these problematic age determinations were obtained for small, epigenetic ice wedges only several decimeters wide that belong to the Kuchchugui Suite (Table 1, samples 7 and 12); these were probably contaminated by ion exchange between small ice wedges and the more concentrated pore ice in the surrounding permafrost deposits. Electrical conductivity transects through such ice wedges show clear and symmetric increases toward the margins [Meyer *et al.*, 2002].

[29] Extrapolating local chronologies to their base ages, as shown in Figure 3, provides an estimate of the range of the assumed $^{36}\text{Cl}/\text{Cl}$ surface input. For all the sites but one, this input falls within a factor of two of 0.3×10^{-12} . The single exclusion is of the much higher values from the Bol'shoy Khomus

Yuryakh River site, though the two measurements made there are not enough for definite conclusions.

[30] No sizable fractionation of the chlorine isotope ratio in the water phase during transport from the surface to ice wedges could be caused either by physical processes like evaporation/condensation or by simple chemical binding. Nonconstant input of sea-borne chloride could produce variations in $^{36}\text{Cl}/\text{Cl}$, but they would be negatively correlated with the chloride concentration. From the geological point of view such anomalous $^{36}\text{Cl}/\text{Cl}$ ages could result from lateral ion exchange between ground ice and the surrounding frozen sediment as water migrated along concentration gradients. Assuming an approximate exchange rate of 10 cm per 10,000 years, small ice wedges are more sensitive to such exchange than are larger ones. Chemical analysis of pore ice from similar deposits showed certain layers with very high chloride concentrations (100–3000 ppm) and high electrical conductivity ($1-10 \text{ mScm}^{-1}$) (H. Meyer, AWI Potsdam, personal communication, 2009). A possible explanation was the high aridity and evaporation in the previous period, which resulted in increased ion concentrations in near-surface soils due to repeated freezing-thawing cycles. Strong evaporation due to high continentality is a rather common climate phenomenon in east Siberia today, with potential evapotranspiration approximately twofold higher than real precipitation [Wetterich *et al.*, 2008b; S. Rivas-Martínez, Global bioclimatics data set, Phytosociological Research Center, Madrid, Spain, available at <http://www.globalbioclimatics.org>]. In addition, there are several paleoenvironmental records of stronger continentality in the study area during the middle and late Pleistocene [Andreev *et al.*, 2004, 2009; Kienast *et al.*, 2005, 2008; Wetterich *et al.*, 2009].

[31] The enhanced ionic composition of surrounding groundwater could be transmitted to postsedimentary epigenetic ice wedges, leading to the false dates. Another process that could be important is thawing of ground ice during warming periods like the Eemian interglacial as described by Wetterich *et al.* [2009] for both coasts of the Dmitry Laptev Strait. Such a thermokarst-related process could result in remobilization of a highly concentrated soil solution and its intrusion into older ground ice bodies.

5. Summary and Conclusions

[32] $^{36}\text{Cl}/\text{Cl}$ measured in ground ice provides valuable information about the timing of permafrost

formation. For the first time, previously unknown permafrost ages were estimated for the vast territory of the eastern Arctic. Except for a few samples, the age of all ice wedges and segregation ice syngenetically formed in cryochrones, demonstrating a good correlation with expected ages as confirmed by other geochronological methods and stratigraphic evidence. Several local chronologies were constructed for syngenetic middle and late Pleistocene ground ice samples from northeast Siberia. Because the lifetime of ^{36}Cl is 434,000 years, the method is more applicable to old (Saalian) ice wedges than to young (Weichselian) ice wedges. More detailed cryolithological (i.e., ice texture) and chemical analyses, such as major ion and trace element concentrations and stable isotope ratios, are necessary to refine this method for determining ice wedge chronology as well as contamination patterns. The results of this study suggest that the reliability of dating permafrost using $^{36}\text{Cl}/\text{Cl}$ ratios depends on the size and on the formation type of the sampled ground ice. In this context syndimentary- (syngenetically) formed large ice wedges are preferred over post-sedimentary- (epigenetically) formed wedges, in order to avoid the influence of brines within surrounding deposits on the initial (precipitation-controlled) $^{36}\text{Cl}/\text{Cl}$ ratio in the ice.

[33] The main conclusion is that the chlorine level and $^{36}\text{Cl}/\text{Cl}$ ratios in the Yedoma Suite, which formed in Weichselian cryochrones (12–15 to 50 thousand years ago), may serve as a zero point (or base) for dating Ice Complex sequences formed in middle and early Pleistocene cryochrones under more or less similar climatic conditions, including precipitation.

[34] The systematic changes of the Holocene samples require additional studies, and the observed change in $^{36}\text{Cl}/\text{Cl}$ ratios during the late Pleistocene to Holocene transition requires additional explanation. Such studies should be connected with comparable analyses of Northern Hemisphere glacier ice as well as of precipitation in the catchment area.

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